COMPONENT PART NOTICE

THIS PAPER IS A COMPONENT PART OF THE FOLLOWING COMPILATION REPORT:

TITLE: Ceramic Transactions. Volume 21. Proceedings of the Symposium on Microwave Theory and Application in Materials Processing Annual Meeting of the American Ceramic Society (23rd) Held in Cincinnatti, OH on April 29-May 3, 1991.

TO ORDER THE COMPLETE COMPILATION REPORT, USE AD-A253 631.

THE COMPONENT PART IS PROVIDED HERE TO ALLOW USERS ACCESS TO INDIVIDUALLY AUTHORED SECTIONS OF PROCEEDING, ANNALS, SYMPOSIA, ETC. HOWEVER, THE COMPONENT SHOULD BE CONSIDERED WITHIN THE CONTEXT OF THE OVERALL COMPILATION REPORT AND NOT AS A STAND-ALONE TECHNICAL REPORT.

THE FOLLOWING COMPONENT PART NUMBERS COMPRISE THE COMPILATION REPORT:

AD#: AD-P007 716 thru AD#: P007 783

AD#: _____ AD#: _____

AD#: _____ AD#: _____

MICROWAVE PLASMA SINTERING OF ALUMINA

Mary P. Sweeney* and D. Lynn Johnson
Northwestern University
Department of Materials Science and Engineering
Evanston, IL 60208-3108

ABSTRACT

A tunable single mode (TM_{012}) microwave cavity was designed to initiate and sustain a plasma for sintering ceramic materials. Almost 100% of the incoming microwave energy could be coupled to the plasma under optimal tuning conditions. An optical fiber thermometer was used to continuously monitor the temperature of alumina specimens during plasma sintering. The sintering temperature and resulting density of the specimens were dependent on gas content at constant net power absorption.

INTRODUCTION

The history of plasma sintering dates back to 1968, when Bennett *et.al.* first reported developing a 2.45 GHz microwave-induced plasma heat source for sintering ceramics.[1] Alumina compacts were found to sinter more rapidly in a plasma and had higher densities, greater strengths and finer grain sizes than conventionally sintered specimens. Evidence of enhanced densification in a plasma also was observed.[2] Specimens that were partially sintered in a discharge showed a negligible increase in densification upon further heating in a conventional furnace at the same temperature. However, densification would continue when the same specimen was reimmersed in the plasma, and its final density would be equivalent to that of specimens that were plasma sintered continuously for the same amount of time. These results suggest that a plasma sintering environment may be more than a simple heat source.

---

*Now at Raytheon Co., 50 Apple Hill Drive, Tewksbury, MA 01876

Extensive work in the field of plasma sintering has been conducted for over ten years at Northwestern University. Microwave applicators and RF and DC hollow cathode plasma devices have been used to generate plasma discharges. Rapid densification rates and anomalous heating characteristics were reported.[3-5] Knowlton first showed that the final densities of RF plasma sintered specimens were strongly related to the amount of dopant gas present during sintering, whether directly added to the plasma or injected into the plasma by desorption from the surface of the ceramic powder particles.[6] Chen further quantified these results and reported that outgassed alumina specimens did not show any significant amount of shrinkage in a 100% argon plasma, but could be heated to sintering temperatures when small amounts of polyatomic gases such as N_2, O_2, H_2 and H_2O were added to the argon.[7] The net power consumption of the plasma could not be accurately determined for these experiments.

The present work was undertaken to further investigate the effects of plasma composition on microwave induced plasma heating of alumina. A tunable single mode (TM_{012}) resonant cavity that had on-line power monitoring and tuning capabilities was designed and implemented for the microwave plasma sintering studies. It has been noticed that a microwave discharge takes on a shape related to the filled pattern of its electromagnetic mode.[8] The TM_{012} mode was chosen because the electric field lines run parallel to the coaxial plasma tube.[9]

EXPERIMENTAL

The single mode (TM_{012}) tunable cylindrical resonant cavity used as the microwave plasma sintering apparatus was designed in accordance with the descriptions published by Asmussen.[8] A schematic representation is shown in Figure 1. The adjustable movable plate short of the cavity served two purposes: tuning the TM_{012} mode and improving impedance matching with the plasma. A coaxial launch probe assembly was used to transmit 2.45 GHz microwave power into the cavity. Both the insertion depth of the probe into the cavity and the position of its short circuit could be individually adjusted to optimize tuning of the system. The 40 mm I.D. fused quartz plasma tube was cooled by compressed air and the outer brass walls were water cooled. A microwave power directional detector was used to continuously monitor power absorption into the cavity.

The alumina specimen configurations were used in this study were 4 mm diameter x 150 mm rods and thimble shaped specimens, 8 mm ID, 11 mm OD and 10 mm high. All specimens (30 m^2/g alumina* with 3% polyvinyl butyral binder) were isostatically pressed and pre-sintered at 650°C in air to burn out the binder. Both types of specimens were held stationary in the cavity, with the rods extending

* Baikowski CR30, Baikowski International Corp., Charlotte, NC 28210

along most of the length of the cavity and the thimbles rested inverted in the center of the plasma on the end of a single crystal sapphire tube*. An optical fiber thermometer** (OFT) lightpipe sensor was inserted into the interior of the thimble to provide temperature readings.

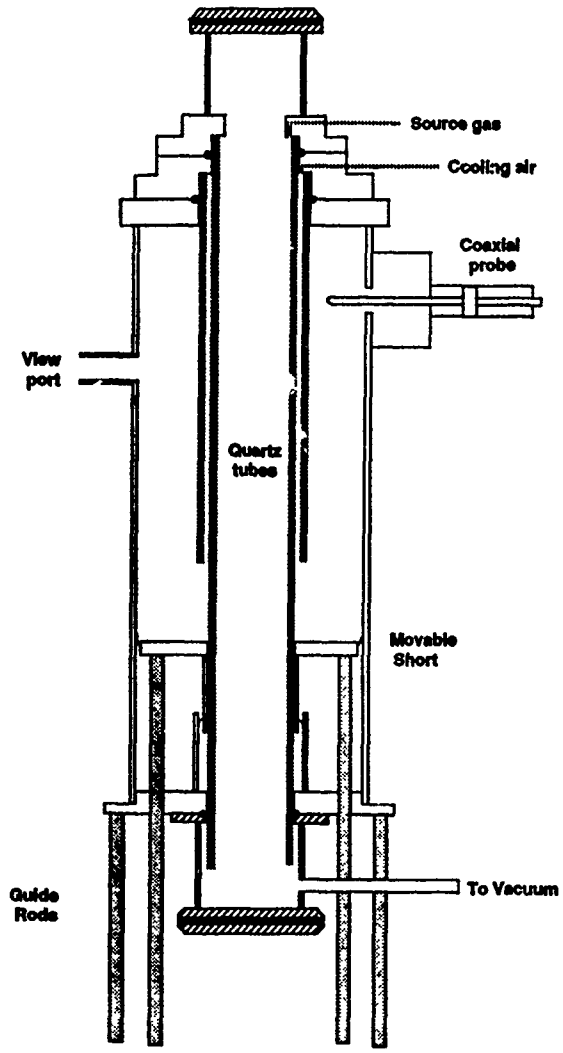

Fig. 1. Schematic cross section of the TM_{012} plasma sintering cavity.

---

* Kindly supplied by R.D. Bagley, Corning Glass Works, Corning, NY 14830

** Accufiber, Inc., Vancouver, WA 98661

The sintering runs for all specimens began by subjecting them to a low power argon plasma to drive off adsorbed gases from the powder particle surfaces without causing sintering. Gas composition and flow rates were adjusted and the power and pressure were brought up to their working levels to initiate sintering.

The alumina rods were plasma sintered with varying amounts of nitrogen or oxygen in argon. The sintering conditions, which were constant for all runs, were 475 W net power absorbed, 14 kPa total pressure, 10 ml/min STP total gas flow rate, and 5 minute soak. The diameters of the rod specimens were measured before and after sintering as a function of position in the cavity. Using the OFT, the same time/temperature profile was reproduced under different plasma gas conditions for the thimble experiments. Here the flow rate was fixed at 2.5 ml/min STP at a pressure of 15 kPa. The final densities of the specimens were determined by Archimedes' method.

RESULTS AND DISCUSSION

Initial experiments showed that under optimal tuning conditions essentially 100% of the forward power could be coupled to the plasma. Certain plasma operating conditions resulted in the melting of quartz and alumina after tuning.

The normalized final shrinkage of plasma sintered alumina rods as a function of position in the cavity is shown in Figures 2 and 3. Rods sintered in a 100% argon plasma showed no evidence of densification with the same net power that caused shrinkage with diatomic molecules present. As the overall content of diatomic gas in the source gas was raised, the shrinkage of the rods increased. The length of the plasma hot zone was approximately constant regardless of diatomic gas content. The enhanced heating effects may be attributed to the additional heating modes of diatomic gases (atomic recombination). The maximum temperature (maximum shrinkage) position was located in line with the coaxial probe, rather than at the center of the cavity as expected for the undistorted TM_{012} mode.

Figures 4 and 5 show the relative density and normalized shrinkage of the sintered rods as a function of diatomic gas content. At low percentages, oxygen provided greater densities and shrinkage rates than nitrogen for the same volume content and same net power. At higher levels the densification effects of both gases converged.

Figures 6 and 7 show the time-temperature profiles of thimble specimens sintered in a variety of plasmas. A summary of the operating conditions and resulting thimble densities can be found in Table 1. As expected, more net power was required to bring the lower diatomic gas levels to the same temperatures as for the higher quantities. Plasma source gases doped with nitrogen required greater overall net power levels to reach the same temperature as those doped with oxygen. A significant increase in density was seen with the 50% nitrogen sintered thimbles.

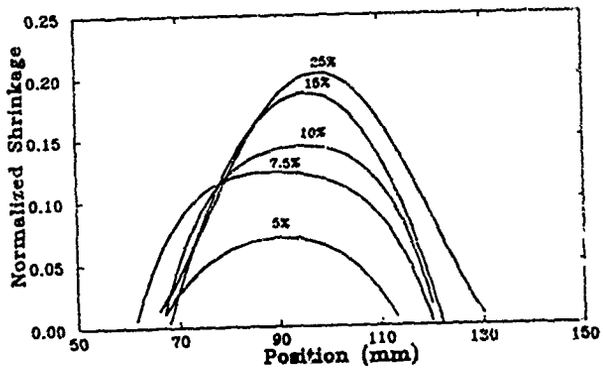

Fig. 2. Normalized diametral shrinkage as a function of distance from the top of the cavity. Indicated are volume percent levels of oxygen in an argon carrier gas.

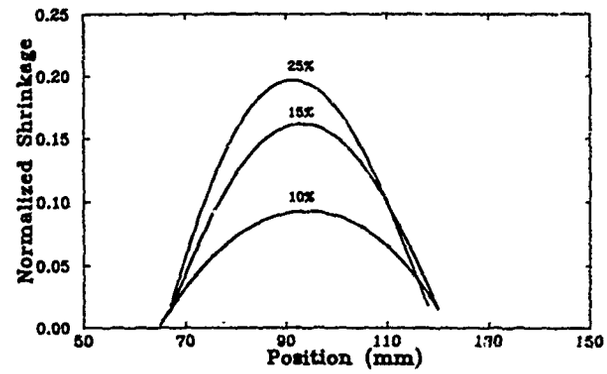

Fig. 3. Normalized diametral shrinkage of plasma sintered alumina rods as a function of distance from the top of the cavity. Indicated are volume percent levels of nitrogen in an argon carrier gas.

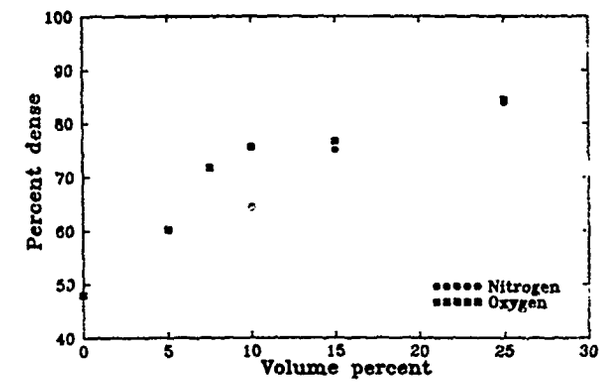

Fig. 4. Maximum density of alumina rods sintered in the plasma as a function of diatomic gas content. Data of Figures 2 and 3.

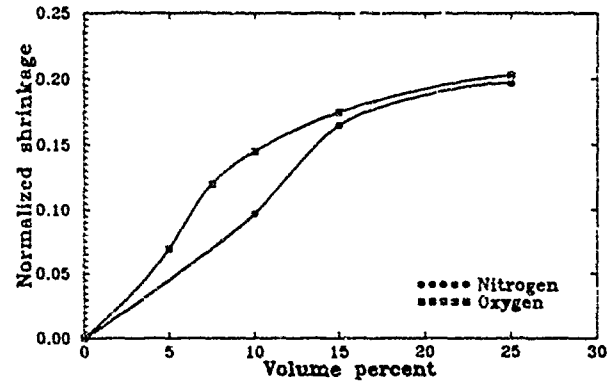

Fig. 5. Normalized maximum diametral shrinkage of plasma sintered alumina rods as a function of diatomic gas content.

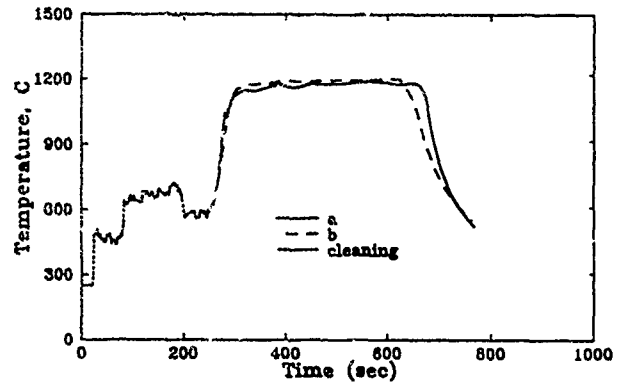

Fig. 6. Indicated temperature-time profiles of alumina thimble specimens sintered under different oxygen gas contents. Label letters refer to Table 1. Low power was applied first to remove adsorbed gases from the specimen.

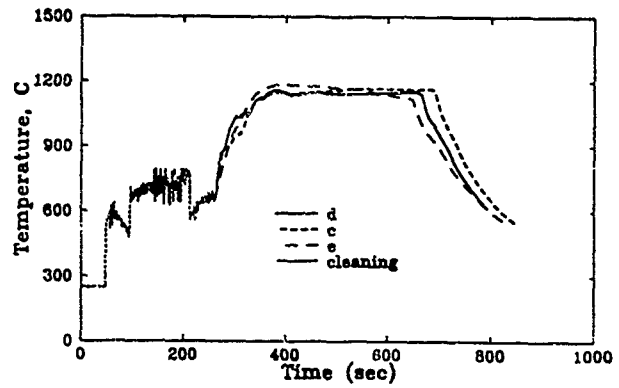

Fig. 7. Indicated temperature-time profiles of thimble specimens sintered under different nitrogen gas contents. Label letters refer to Table 1.

Table 1. Time-Temperature Thimble Runs

Run ID	Volume percent of Additive Gas	Net Power (watts)	Relative Density
a	50% O_2	600	79.1%
b	10% O_2	880	84.2%
c	50% N_2	870	94.9%
d	50% N_2	820	93.8%
e	10% N_2	1180	83.8%

After the present work was completed, Hansen observed that the OFT reads a temperature that is biased to significantly lower values because of translucency of the thimble specimens at the wavelength of radiation sampled by the OFT.[10] While the temperatures shown in Figures 6 and 7 are substantially below the true temperatures, it is reasonable to suppose that the error is approximately the same for all specimens since they do not differ greatly in density (and therefore translucency).

CONCLUSIONS

The single mode (TM_{012}) tunable resonant cavity was shown to be a viable means for microwave-induced plasma sintering. Advantages of this system include real-time monitoring of the net power absorbed by the plasma and tuning capabilities to optimize power coupling. Sintering studies with alumina showed that the previously recognized enhanced sintering characteristics in diatomic gases were not a function of power absorbed by the plasma, but rather depended upon the chemical composition of the plasma.

ACKNOWLEDGEMENTS

This work was supported by the National Science Foundation Grant DMR-8719077, and made use of Central Facilities supported by the National Science Foundation through the Northwestern University Materials Research Center, Grants Nos. DMR 85-20280 and 88-21571. The contributions of Prof. M.E. Brodwin, Department of Electrical Engineering and Computer Science, particularly the design of the launch probe, are gratefully acknowledged.

REFERENCES

1. C.E.G. Bennett, N. A. McKinnon, and L. S. Williams, "Sintering in Gas Discharges," Nature, 217, 1287-88 (1968).

2. C.E.G. Bennett and N. A. McKinnon, "Glow Discharge Sintering of Alumina," in Kinetics of Reactions in Ionic Systems, Edited by T. J. Gray and V. D. Frechette, Plenum Press, New York, 1969; pp.-408-12.

3. Joung Soo Kim and D. Lynn Johnson, "Plasma Sintering of Alumina," Am. Ceram. Soc. Bull., 62 [5] 620-22 (1983).

4. D. L. Johnson, V. A. Kramb, and D. C. Lynch, "Plasma Sintering of Ceramics," in Emergent Process Methods for High-Technology Ceramics, Vol. 17, Edited by R. F. Davis, H. Palmour III, and R. L. Porter, Plenum Publ. Corp., 1984; pp. 207-211.

5. E. L. Kemer and D. L. Johnson, "Microwave Plasma Sintering of Alumina," Am. Ceram. Soc. Bull., 64 [3] 1132-36 (1985).

6. J. M. Knowlton, "Radio Frequency Plasma Sintering of Alpha-Alumina Doped with Various Oxides," MS Thesis, Northwestern University, 1985.

7. M. Y. Chen and D. Lynn Johnson, "Effects of Additive Gases on Radio Frequency Plasma Sintering of Alumina," J. Mat. Sci., accepted for publication.

8. J. Asmussen, Jr., R. Mallavarpu, J. R. Hamann, and H. C. Park, "The Design of a Microwave Plasma Cavity," Proceedings of the IEEE 1, 109-117 (1974).

9. J. Asmussen and R. Garard, "Precision Microwave Applicators and Systems for Plasma and Materials Processing," in Materials Research Society Symposium, Vol. 124, (Materials Research Society, 1988), pp. 347-352.

10. J. D. Hansen, private communication, 1991.